# General.

We would like to thank the anonymous Referee #3 for providing comments to improve and clarify our manuscript. We will revise the text by fully taking the comments into account. Please find our responses to the specific comments and questions below. Our response is written in bold. The revised parts of the manuscript are highlighted in bold red.

# 5 Comments of Referee #3 and our responses to them

### **General comments**

Muller et al., developed three approaches to construct the CH4 vertical profiles, and then use them to calculate the XCH4. They then show how these observation-based XCH4s can be used to investigate the seasonal variation in CH4 and to evaluate the satellite observations of CH4. This study involves a large amount of data, including airborne and ship

10 measurements, satellite observations, and model simulations. This manuscript is well-organized and is within the scope of AMT. I recommend its publication after the authors address the following comments:

### **Specific comments**

### Comment 1

Sampling bias is a big concern as the ship observations are 6 + 4 days and the airborne observations are 2+1 days each month. The uncertainty arising from limited measurements is not well covered in Sect 3.3.1.

### Response

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Thank you for pointing this out. The uncertainty due to the limited number of in situ data is currently the major drawback of our approach. We added the following explanation at the beginning of chapter 3.3:

- 20 Lines 241–246: There are two uncertainty sources. The first uncertainty source arises from the limited number and spatiotemporal distribution of in situ data within the latitude-longitude boxes of each month. Therefore, the data may not always represent the monthly averaged CH<sub>4</sub> concentration within the area of interest accurately. However, in the near future, the number of in situ data will increase and the spatial distribution expands as discussed in chapter 5. The second source of uncertainties in the obs. XCH<sub>4</sub> (simple, blended) are caused by the CH<sub>4</sub> profile construction: a) the inter- and
- 25 *extrapolation of the in situ data in the troposphere, b) the tropopause height, and c) the modelled stratospheric column.*

# Comment 2

Figure 2 includes a lot of information. I would add observation-based profiles from 3 approaches in different colors, instead of showing them in one symbol.

## 30 Response

We agree that Fig. 2 was not clear. The original Fig. 2 showed the interpolated observation-based profile of approach 2. The other two approaches were not specifically shown. Therefore, we revised Fig. 2 by adding subfigures for each approach and added references to each subfigure to the text as follows below.



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**Figure 2:** Construction of the observation-based CH<sub>4</sub> profile (blue) obtained by using ship and aircraft data (yellow) together with model results (green), and the interpolation onto the pressure grid of the satellite retrieval (red) for approach 1 a), approach 2 b), and approach 3 c). The example is obtained at the latitude  $30-40^{\circ}$  N, in March 2015.

Line 223: Approach 1 is the adaptation of the approach of Müller et al. (2021) (Fig. 2a).

40 Lines 226–228: Approach 2 is the addition of JMA aircraft data to the mid troposphere (Fig. 2b). We linearly interpolate between the extrapolated ship data, and both aircraft data. In approach 3, we fill in model results between the aircraft data of JMA and CONTRAIL of approach 2 (Fig. 2c).

# Comment 3

**45** Figures 3 and 4 are very difficult to read. It is also unclear where is the gap in the dataset. In Figure 3, the linear regression fitted line is not in the legend and has the same color as blended obs/ XCH4.

## Response

In accordance with a similar comment of referee #1, we revised Fig. 3 and 4 by only showing the 16 ppb uncertainty range of approach 3 (blended obs. XCH<sub>4</sub>), and ACTM<sub>XCH4</sub> as grey area. Furthermore, we removed the comparison

50 with the TCCON stations from Fig. 3 to make the comparison of the approaches clearer. Instead, we added a new Fig. 4 which only shows results of approach 3 in comparison with those of the two TCCON stations. The linear fit is added to the legend in a color different to that of the blended XCH<sub>4</sub>.

Data points are connected by straight lines. In the revised Figures with reduced information, the data gabs become clearer, seen as long straight lines between the markers.

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We also revised Fig. 5 (now Fig. 6) and Fig. A3 (now Fig. A4) in order to have the same color depth. In addition, we revised the caption of the new Fig. 6 and new Fig. A4 by adding the description of the uncertainty range:

Lines 443–445: Figure 6: Temporal variation of the blended obs. XCH4 (ACTMxCH4, black) in comparison with GOSAT XCH4 retrievals
from NIES (orange), RemoTeC (blue), and OCFP (green) at the latitude range 30–40° N (a) and 20–30° N (b). The grey area is the 16 ppb uncertainty of the blended obs. XCH4.

Lines 511–513: Figure A4: Temporal variation of the blended obs.  $XCH_4$  ( $ACTM_{XCH4}$ , black) in comparison with GOSAT  $XCH_4$  retrievals from NIES (orange), RemoTeC Heidelberg (HD) (magenta), RemoTeC SRON (blue), and OCFP (green) at the latitude range  $g1 = 30-40^{\circ} N$  (a) and  $g2 = 20-30^{\circ} N$  (b). The grey area is the 16 ppb uncertainty range of the blended obs.  $XCH_4$ .



**Figure 3:** Temporal variation of monthly averaged XCH<sub>4</sub> obtained by approach 1 (simple obs. XCH<sub>4</sub>, green), approach 2 (obs. XCH<sub>4</sub>, orange), and approach 3 (blended obs. XCH<sub>4</sub>, black) at the latitude range 30–40° N (a) and 20–30° N (b). The uncertainty ranges are 22 ppb, 20 ppb, and 16 ppb for approach 1, 2, and 3 respectively. Only the 16 ppb uncertainty range of approach 3 is shown as grey area. Uncertainty ranges of the other approaches are not shown for readability.



Figure 4: Temporal variation of monthly averaged XCH<sub>4</sub> obtained by approach 3 (blended obs. XCH<sub>4</sub>, black), and from the TCCON station in Saga (green) and Tsukuba (orange) at the latitude range 30–40° N (a) and 20–30° N (b). The grey area is the 16 ppb uncertainty range of approach 3; error bars are the standard deviations of TCCON. Also shown is the linear least-square regression (deep blue line) with a 90% confidence interval on the slope and intercept (deep blue dashed line) of approach 3.

#### ← ACTM<sub>XCH4</sub> → CAMS<sub>XCH4</sub> → CAMSinv<sub>XCH4</sub>



Figure 5: Comparison between the blended obs. XCH<sub>4</sub> (approach 3) derived from CH<sub>4</sub> profiles using the MIROC4-ACTM (ACTM<sub>XCH4</sub>, black), CAMS (CAMS<sub>XCH4</sub>, green), and CAMSinv (CAMSinv<sub>XCH4</sub>, orange) for the stratospheric column at the latitude range 30–40° N
 (a) and 20–30° N (b). The uncertainty range of all results is 16 ppb. The grey area is uncertainty of ACTM<sub>XCH4</sub>. Uncertainty ranges of the other results are not shown for readability.

### Comment 4

Line 154: Why do you choose the data that only assimilates NOAA surface observation? How is it different from assimilation using both NOAA surface observations and GOSAT observations?

### Response

Our aim is to provide a reference dataset for satellite validation as complement to other networks like TCCON. The model used in our approach has to be independent from the satellite retrieval, which we want to evaluate. Therefore, we do not use data which assimilates GOSAT data. However, the assimilation of precise in situ data improves the

## 90 accuracy of model calculations.

### We clarified the sentence as follows:

Lines 155–156: We choose datasets which assimilate NOAA surface observations, but not GOSAT observations to ensure that the model results in our approach are independent from the satellite we aim to validate.

## **95** Line 367-370: Why the increasing trend in XCH4 is larger between 20-30N than 30-40N.

## Response

The first thing to note is that fewer data at 30-40° N as compared to 20-30° N might have cause an artificial difference in the growth rate between the latitude ranges as mentioned in lines 411–412.

Besides this, the growth rate is influenced by the complex interaction of various factors, especially by anthropogenic 100 CH<sub>4</sub> emissions, the availability of OH radicals as a primary oxidant for methane, and atmospheric circulation patterns. To confirm that there is a real difference in the growth rate of the neighbouring latitude ranges, more comprehensive analysis is needed.

However, given a lower growth rate in the northern latitude range, combined with a higher similarity of our obs. XCH<sub>4</sub> with those XCH<sub>4</sub> influenced by the Asian emission outflow at Saga (chapter 4.1), we can suggest that the

105 interaction between various anthropogenic emissions might have led to higher OH concentrations, consequently, CH<sub>4</sub> removal rates near to the Japanese East coast (Fig. 1), and therefore causing a slower annual growth rate. Another explanation can be the decreasing trend of CH<sub>4</sub> emissions from Japan related to policy changes (Ito et al., 2023).

## We added the following explanation:

- 110 Lines 411–417: It is noted that limited and uneven sampled in situ data during each month might cause an artificial difference of the growth rates between the latitude ranges. However, given a lower growth rate at the higher latitude range combined with a higher similarity of the blended obs. XCH4 with those XCH4 influenced by the Asian emission outflow at Saga (chapter 4.1), we can suggest that the interaction between anthropogenic emissions might have led to increased OH concentrations, consequently higher CH4 removal rates near to the Japanese East coast (Fig. 1), and therefore causing a slower annual growth. Or, it might indicate that, compared to 20–30° N, the higher latitude range is
- 115 therefore causing a slower annual growth. Or, it might indicate that, compared to 20–30<sup>•</sup> N, the higher latitude range is affected by the decreasing trend in CH<sub>4</sub> emission from Japan (Ito et al., 2023).

Lines 643–644: Ito, A., Patra, P. K., and Umezawa, T.: Bottom-Up Evaluation of the Methane Budget in Asia and Its Subregions, Global Biogeochemical Cycles, 37, https://doi.org/10.1029/2023gb007723, 2023.

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### Other corrections made

Lines 306–308: *For that, we interpolated the MIROC4-ACTM data with its higher resolved pressure grid on that of the CAMS and CAMSinv data, respectively (section 2.3).* 

Lines 538-539: CAMSinv data were provided by CWO.

125 CWO was added under Author contribution beside being not a co-author. We corrected this.

Lines 608–610: Copernicus Climate Change Service, Climate Data Store: Methane data from 2002 to present derived from satellite observations, Copernicus Climate Change Service (C3S) Climate Data Store (CDS), https://doi.org/10.24381/cds.b25419f8, 2018, accessed on 17 May 2023.

 Lines 695–697: NIES GOSAT Project: Release Note of Bias-corrected FTS SWIR Level 2 CO2, CH4 Products

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 (V02.95/V02.96)
 for
 General
 Users",

 https://data2.gosat.nies.go.jp/doc/documents/ReleaseNote\_FTSSWIRL2\_BiasCorr\_V02.95-V02.96\_en.pdf, 2020, revised
 2021, accessed on 21 April 2023.

Lines 812–815: Yoshida, Y., Someya, Y., Ohyama, H., Morino, I., Matsunaga, T., Deutscher, N. M., Griffith, D. W. T., Hase, F., Iraci, L. T., Kivi, R., Notholt, J., Pollard, D. F., Té, Y., Velazco, V. A., Wunch, D.: Quality evaluation of the column-

135 averaged dry air mole fractions of carbon dioxide and methane observed by GOSAT and GOSAT-2, Scientific Online Letters on the Atmosphere (SOLA), 19, 173–184, https://doi.org/10.2151/sola.2023-023, 2023.

## References

Ito, A., Patra, P. K., and Umezawa, T.: Bottom-Up Evaluation of the Methane Budget in Asia and Its Subregions, Global Biogeochemical Cycles, 37, https://doi.org/10.1029/2023gb007723, 2023.